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must be high and M' low. If C' is at G' the whole of each component beam may be caught and passed through the respective shanks of the U-tube. The fringes are strong, easily found and large, so that the center of ellipses is not far outside of the field of the telescope.

Finally if the connecting tube p is nearly horizontal when in place, the fringes are usually found at about the same position of the micrometer (at M') after the liquid is introduced into the U-tube.

Experiments were also made with this apparatus. Displacement interferometers in which the rays do not retrace their respective paths have an important special property which I wish to accentuate in conclusion. If either opaque mirror is displaced on its micrometer normal to itself or if a plate compensator rotates in one beam on a vertical axis, the center of ellipses moves parallel to the length of the spectrum. If however the plate compensator rotates on a *horizontal* axis, the center of ellipses moves nearly *transversely* to the length of the spectrum. The phenomenon is quite sensitive. To this result I shall return in a succeeding note, in connection with the development of the Jamin design for displacement interferometry.

The present note will be presented in more extended form in a report to the Carnegie Institution of Washington.

ATTEMPT TO SEPARATE THE ISOTOPIC FORMS OF LEAD BY FRACTIONAL CRYSTALLIZATION

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Although the complete inseparability of isotopes by chemical means has been frequently asserted, the evidence on which this assertion is based has always seemed insufficient. The methods used have been fractional crystallization and precipitation, but these processes have seldom been carried out more than ten times in a particular case, and frequently six or seven crystallizations have been thought a sufficiently thorough test of inseparability. A search of the literature revealed only one investigation, that of Radiothorium and Thorium by McCoy and Ross,[1] where as many as one hundred repetitions of a given process had been made.

It seemed worth while, therefore, to apply to the important generalization of Fajans, Russell, Fleck and Soddy a more searching test carrying the fractionation further, and using as a criterion of success not only the measurement of radioactivity but also the determination of atomic weight.

The lead from Australian carnotite, with which the kindness of Mr. S. Radcliff and Mr. E. R. Bubb had supplied us and which had been used in previous atomic weight and density determinations in this laboratory,[2] seemed well suited to our purpose.

About 1 kilogram of this lead was converted into nitrate and used for the fractional crystallization. Lead nitrate crystallizes from aqueous solution in compact, large octahedra which can be very completely freed from mother liquor merely by drainage, and come rapidly to equilibrium with the mother liquor.

The whole of the nitrate was dissolved in distilled water and evaporated until a crust began to form. This was called fraction 1. After cooling to room temperature the mother liquor was poured off and formed fraction 2. To the crystals from fraction 1 was added sufficient water to dissolve them, and the solution was called fraction 3.

Fractions 2 and 3 were evaporated to saturation and cooled as fraction 1 had been, the liquor from 2 forming fraction 4. Fraction 5 was made by adding the liquor from 3 to the crystals from 2, and water was added to the crystals from 2 to form fraction 6. This process was continued until, as occurred first with fraction 16, one of the end fractions became too small to work with conveniently and was consequently added entire to the mother liquor from fraction 17 to form fraction 22. This expedient was resorted to wherever the end fractions became inconveniently small. In all, seventy-five courses or tiers of fractions were obtained, most of the last of which contained eighteen fractions to a tier. The number of effective crystallizations was 904.

The crystallization was carried out entirely in porcelain, at first in large evaporating dishes, then, as the fractions became smaller, in casseroles covered with watch glasses. The hot casseroles were usually set in evaporating dishes full of cold water, which was frequently changed to hasten cooling.

Centrifugal drainage was not used because, as determined by calculation, the gain in effectiveness of separation would have been less than the disadvantage caused by the increased time necessary to treat the precipitates centrifugally.

Purification.—Two samples, one from the more soluble and one from the less soluble end of the final row of crystallizations were treated in an identical manner. Chiefly to remove the large amount of silica which had dissolved from the casseroles during the fractionation, the crystals were first electrolyzed in warm concentrated solution between platinum wires. The well formed flat leaflets of lead were removed from time to time, with glass rod, rinsed with a stream from a wash bottle, and preserved in quartz dishes until all the metal had been deposited from the solution. The crystal were further washed from time to time, and all washings returned to the electrolysis bath, which was gradually reduced in volume as the concentration of lead became smaller. The lead crystals were then dissolved in pure nitric acid, and the excess of acid was evaporated. The crystal of lead nitrate were dissolved in the purest water, filtered through a platinum Gooch crucible, and precipitated by the addition of a large excess of redistilled nitric acid and cooling with ice. The crystal meal thus obtained was freed from adhering acid in the platinum centrifuge, washed centrifugally with purest concentrated acid and redissolved in the least possible amount of pure boiling water.

Five precipitations with centrifugal drying were made in this way. The crystals from the fifth crystallization were placed in a quartz dish and dissolved. From this solution lead chloride was precipitated by passing in pure hydrochloric acid gas made by warming the purest acid of commerce. The chloride thus obtained was dried in the centrifuge, washed with cold water, and redissolved in boiling water. Four crystallizations as chloride followed. A little hydrochloric acid was added to the solution each time to prevent the formation of basic salt and to render the chloride less soluble. The last crystals, after centrifugal washing and drying were preserved in an exhausted desiccator over fused potash. Nothing but platinum and quartz vessels were used.

The Determination of Atomic Weight.—For these determinations the usual procedure was employed. About five grams of the dried crystals were placed in a platinum boat, which with its container had been carefully weighed. The boat was gradually heated in a quartz tube attached to the usual 'bottling apparatus,' in a current of pure dry hydrochloric acid gas. After nearly all the residual water had been driven from the crystals, the temperature was raised so as to fuse the salt, which was then cooled as quickly as possible in a current of pure dry nitrogen prepared by the Wanklyn process.

After the salt had cooled, and the nitrogen had been replaced by dry air, the boat was pushed into the weighing tube, bottled, and reweighed.

The boat and salt were next heated at 80°–90° on an electric stove in a glass flask containing about a liter of water to which enough pure nitric acid had been added to prevent the formation of basic salt, until the lead chloride had all dissolved. As usual[3] a small amount of siliceous and carbonaceous residue had to be removed by scrupulously quantitative filtration through a tared Gooch-Monroe crucible. On the average, about twice as much was found in the more soluble fraction as in the other. The weight thus removed was subtracted from the weight of the fused salt. Before filtration the boat had been carefully lifted out and thoroughly washed; and the filtration was collected in the precipitating flask.

A supposedly equivalent weight of silver was next dissolved in pure nitric acid mixed with an equal bulk of water in a flask with a chain of bulbs ground in to the neck, a steam bath being used to heat the solution. More water was then added and the temperature raised until a few seconds' gentle boiling had removed nitrous fumes. The solution having been made up to a liter, it was added very slowly with constant agitation to the lead chloride solution. Actinic light was henceforth rigorously excluded from the precipitated silver chloride. The precipitate was coagulated by violent shaking, usually for fifteen minutes, and allowed to stand at least twenty-four hours, with further occasional shakings, before samples were removed for tests. The end point was found by the customary method by means of the nephelometer, and tested for several days to be certain that it did not change.

Vacuum corrections were applied as usual, and all customary precautions were taken. The results follow.

TABLE 1

ATOMIC WEIGHTS

	ORDINARY LEAD	CARNOTITE A	LEAD B
	207.187	206.426	206.406
	207.187	206.409	206.422
		206.431	206.399
Average.......	207.187	206.422	206.409

This table includes all the analyses of these materials which were completed; any which had become the object of suspicion were rejected at an early stage.

Obviously, if the fractional crystallizations produced any change in the relative concentrations of radium G and lead, this change is very small. The means of the two series of determinations of isotopic lead differ only by about 6 parts in 100,000. This is certainly within the bounds of possible experimental error, though the low 'average errors' of the two sets (0.008 unit in each case) tempt one to assign a meaning to the observed difference. At any rate, we are safe in concluding that assuming the atomic weights of 'radium G' and lead to be 206.0 and 207.2, respectively, no change in concentration greater than 13/1200 or 1.1% has been obtained by nine hundred crystallizations. Even supposing that a real difference in solubility exists, it is evident that complete separation could not be attained (if at all) in less than eighty thousand fractionations.

Estimation of Radioactivity.—The β-ray activity of the lead used indicates that it contains in about 2 parts so-called 'radium G' and 1 part lead, an amount of 'radium D' of the order of 10^{-7} parts. This is betrayed by the steady growth of a penetrating β-ray product (RaE) which comes to practical equilibrium with its parent in about a month after they are separated. Evidently the ionization—if the α-rays from the polonium present are cut off—caused by a weighed amount of the material under constant conditions, is a measure of the concentration of radium D relative to its isotopes 'radium G' and lead. Thus the determination of the activity of the end fractions gives information as to the relation between the solubility of the nitrate of 'radium D' and the mean solubility of the nitrates of 'radium G' and ordinary lead, a relation of great interest because it cannot be tested by atomic weight determinations. This case should afford an especially favorable test of the theory of complete identity.

The method was as follows: To eliminate differences in moisture content, and consequently in absorbtive power, the two samples were kept in the same exhausted desiccator after purification. When sixty-six days had elapsed 10.25 ± 0.03 gram of each sample was weighed into a marked metal dish and tamped with a platinum spatula. The material formed a layer of thickness defined by the expression 0.88 $g/cm.^2$ This is far too thin to give a maximum ionization due to the complete absorption of the β-rays from the lowest layers, but is thick enough so that no great accidental changes in thickness needed to be feared, and the effect of small changes was annulled as described below. The dishes were covered in turn with the same thin sheet of paper and introduced into a grounded aluminum-leaf electroscope which was very kindly loaned by Prof. William Duane. The rate of fall of the leaf was then observed with a stop-watch in the usual manner. All errors arising from differences of potential, differences of stiffness of the leaf in different positions, and inhomogeneity of the scale were eliminated in the usual way. The two dishes were introduced alternately into the electroscope to eliminate the effect of changes in the natural leak, and were thoroughly stirred and repacked between every two measurements to allow for small accidental differences in thickness, and other precautions to cut out extraneous effects and secure exact comparability were employed.

It is believed that the table below, which includes all the observations affected by no known source of error, represents about as good reproducibility as this electroscope can be made to give.

TABLE 2

β-RAY ACTIVITIES

SAMPLE A

11.30; 11.43; 11.69; 11.56; 11.61; 11.47; 11.49; 11.50; 11.41; 11.58 Mean 11.504.

SAMPLE B

11.32; 11.62; 12.06; 11.43; 11.70; 11.34; 11.32; 11.43; 11.47; 11.36; Mean 11.505.

The ionization is expressed in arbitrary units (scale divisions per minute.) The natural leak (which must have been the same for both as they were taken alternately) was found to be 0.50 ± 0.01. The agreement of the averages to one part in ten thousand must be regarded as fortuitous, since their mean probable error is about 0.11 unit. This error, divided by the mean value from which has been subtracted the natural leak may be taken, roughly, as a measure of the accuracy of the determination. Its value is $\frac{11}{1150-50} = \frac{1}{100}$. This work definitely shows, then, that no change in the relative concentration of radium D and its isotopes as great as one per cent has occurred in nine hundred fractionations, and certainly gives no indication that *radium D* could be completely separated from its isotopes by this method even by crystallizing as many as 90,000 times.

If ordinary lead is a mixture of isotopes, this mixture must have been made very long ago while the earth was still in a highly mobile condition (since all the ordinary lead throughout the world seems to have the same atomic weight[4]). Could the composite nature of ordinary lead be proved, the identity of the several samples through geological aeons would form another argument in favor of the inseparability of the constituents.

To the Carnegie Institution of Washington we are greatly indebted for some of the apparatus and material used in this research, which is to be continued in the near future.

Summary.—Lead from Australian carnotite (believed to contain about one part of ordinary lead to two parts of radium G, with a mere trace of radium D) has been fractionally crystallized over nine hundred times as nitrate, and the end-fractions purified.

The atomic weights of the samples so obtained from the crystal and the mother liquor ends of the series respectively agreed within the experimental error of 6 parts in 100,000.

The β-ray activities agreed within the experimental error of 1%.

These observations indicate that the nitrates of Radium D and lead on the one hand and radium G and lead on the other hand could hardly be separated, if at all, by less than 80,000 or 100,000 crystallizations.

Hence one might infer that the molal solubilities of the nitrates are probably essentially identical. The outcome gives strong experimental support for the hypothesis that isotopes are really inseparable by any such process as crystallization.

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[1] *J. Amer. Chem. Soc., Easton, Pa.*, **29**, 1907, (1709).

[2] Richards and Wadsworth, These PROCEEDINGS, **2**, 1916, (505, 694).

[3] Richards and Wadsworth, *J. Amer. Chem. Soc. Easton, Pa.*, **38**, 1916, (2616).

[4] Baxter and Grover, *Ibid.*, **37**, 1915, (1058).

HYBRIDS OF ZEA TUNICATA AND ZEA RAMOSA

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Communicated by E. F. Smith, March, 16, 1917

Zea tunicata and *Zea ramosa* are the two most striking variations or mutations from normal maize. Though usually referred to as agricultural species they seem to deserve a place with the so-called species of Oenothera which have originated by mutation.

The chief characteristic of *Zea tunicata* is that the glumes of the female inflorescence, or ear, are developed so that each seed is entirely enclosed. Associated with this character is a less conspicuous lengthening of the glumes of the staminate inflorescence that results in a thickening of the tassel.

The origin of *Zea tunicata* is not known, but its occurrence in widely separated and isolated regions would indicate that it has originated independently more than once, presumably as a mutation from ordinary maize.

In hybrids with non-tunicate varieties the tunicate character behaves as a dominant, but in our experiments it has never been possible to secure a homozygous tunicate strain. Progenies resulting from the selfing of tunicate plants have always shown segregation into approximately three tunicate plants to one normal.

The tunicate plants in self-pollinated progenies are separable into two classes, one producing typical tunicate ears and thickened tassels like the parent plant, and the other with greatly enlarged tassels containing both staminate and pistillate flowers, and with the ear either aborted or bearing greatly enlarged and usually sterile spikelets. This last class represents approximately one-third of the tunicate plants. Although these plants produce what appears to be normal pollen in the terminal inflorescence the long glumes never open and the pollen is not shed, and we have not been successful in securing selfed seed of this form.